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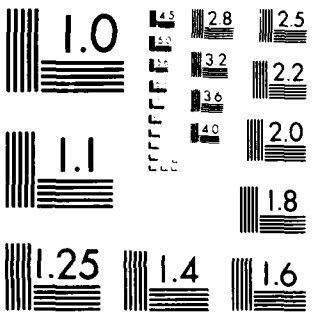

VOLATILITY OF AEROSOLS IN THE WESTERN EUROPEAN ENVIRONMENT

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CONTRACT NUMBER : DAJA45-86-C-0051

4th Interim Report

May 1988

The research reported in this document has been made possible through the support and sponsorship of the U.S. Government through its European Research Office of the U.S. Army. ~~This report is intended only for the internal management use of the Contractor and the U.S. Government.~~

VOLATILITY OF MARITIME AEROSOL OVER THE PERIOD 10 - 11 MARCH 1988

Volatility measurements of ambient aerosol particles were made nearly continuously at Mace Head Field Station over the period 10 - 11 March 1988. A 50 minute temperature cycle was used throughout. The air containing the particles was drawn through the fused quartz tube heated to a temperature of 400°C in about 6 minutes, then allowed to cool for about 44 minutes to a temperature close to ambient and the cycle repeated. A fraction of the particles evaporate and the remainder and/or particle residues then pass through a short section of unheated tubing into a light scattering aerosol counter (a Particle Measuring Systems Model ASASP-X) where their size and number concentration are measured as a function of temperature. The size calibration of Particle Measuring Systems ASASP-X light scattering counter is given in Table 4.

Table 4. Size calibration of Particle Measuring Systems ASASP-X light scattering counter.

	Range 3	Range 2	Range 1	Range 0
Particle diameter intervals (μm)	0.09 - 0.20	0.15 - 0.30	0.24 - 0.84	0.6 - 3.0

An example of Eastern Atlantic aerosol volatility data taken at Mace Head showing aerosol temperature and aerosol concentration for range 3 of the ASASP-X versus (local) time is shown in Fig.5. The measurements represent averages over fairly short time periods (20 s). The modulation of particle concentration induced by heating the aerosol over 50-minute cycles is evident.

The volatility data for the four particle size ranges (see Table 4) is shown for the same period in Fig. 6. The submicron particle concentrations are reduced by heating showing the volatile nature of maritime aerosols. On the other hand the supermicron particles (range 0) appear to be unaffected by heating.

A similar plot as for Fig. 6 is shown in Fig. 7 for the earlier time period from 1745 to 2115 local time. It can be seen that the particle concentration is almost identical for particle ranges 2 and 3. It is also evident that the application of the high temperature causes a greater depletion in number concentration than in the later time period.

This data is replotted showing temperature dependence of maritime aerosol concentration in Fig.8 for five temperature cycles for the later time period for particle counter ranges 3 to 0. The field data is averaged over the entire 250 minute period comprising the five temperature cycles so that only a single fractionation curve results for each particle size range. The resulting average fractionation curves for submicron particles compare favourably with laboratory data for ammonium sulphate (and ammonium bisulphate). This suggests that these materials are a major constituent of the submicron fraction of maritime aerosols. Range 0 particles are largely unaffected by heating. A similar plot is shown in Fig.9 for the earlier time period, averaged over a 150 minute period comprising three temperature cycles. There is favourable agreement between the field measurements and laboratory data using a polydispersion of ammonium sulphate particles.

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Page 1

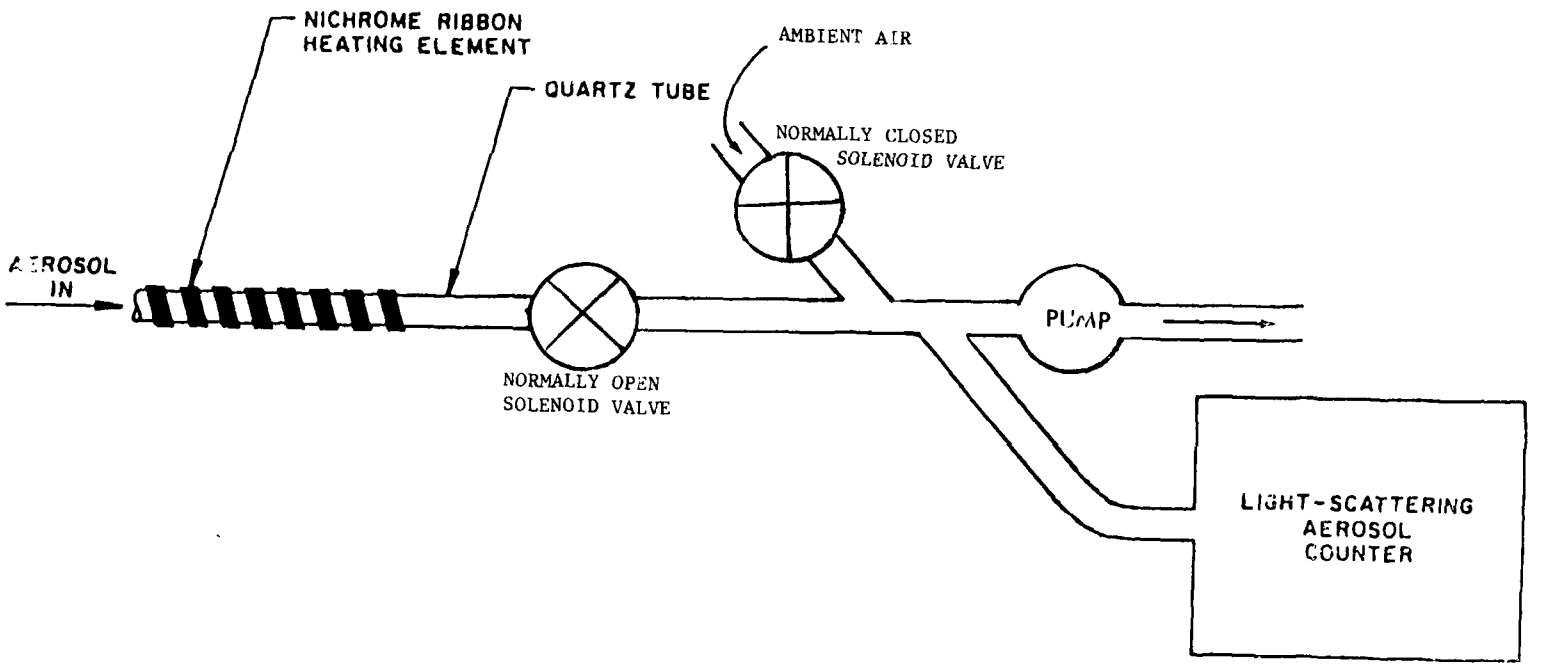

Fig. 1 Schematic of the initial aerosol particle intake setup used to measure particle volatility.

cycling period. Results of the comparisons are given in Table 1. Average losses of 12.1%, 14.0%, 19.6% and 62.8% occur in ranges 3, 2, 1 and 0 for particles passing through the N.O. solenoid valve as compared to a direct flow to the particle counter. This is thought to be mainly due to
(a) a much smaller diameter sampling line in the valve itself;
(b) the presence of a sharp L shaped bend characteristic of the normally closed valve.

A comparison of particle concentration was also made for close to identical sampling line volumes with one line being a straight through line to the ASASP-X whilst the other line was passed through the normally closed (N.C.) solenoid valve. Results of the comparison are shown in Table 2. Average losses of 4.5%, 4.6%, 6.15% and 18% occur in ranges 3, 2 1 and 0 for particles passing through the normally closed valve. The losses are less severe for the N.C. valve since it has a straight through sampling line.

Temperature Probe Tests

Initial field measurements were carried out at an upper temperature of 300°C. For later field tests (see Table 3), the temperature control module was modified to extend the temperature range to 400° (maximum readability for Copper-Constantan thermocouples).

It was found that the presence of the Copper-Constantan thermocouple inside the fused quartz tube reduced particle concentration. Therefore it was decided to place the thermocouple probe on the outside of the quartz tube. Comparative temperature measurements were made for thermocouple probes placed at the "hot-spot" both inside and outside the quartz tube for the upleg and downleg heating cycles. This allowed the inference of the inside temperature as a function of the outside temperature through a fifth order polynomial fit as shown in Fig.2. A plot of the outer thermocouple reading (proportional to a millivolt reading) versus temperature is shown in Fig.3 and Fig.4 for the outer and inner thermocouple probe locations.

Field Measurement Tests

A catalogue of the dates of the various field measurement tests is shown in Table 3. The most recent field data of April 19 - 22, 1988 is yet to be analysed. A preliminary analysis of the earlier data has been made. However it was decided to perform a rigorous analysis first on the March 10 - 12 data since it represents the first continuous data set without the use of the solenoid valves. The thermocouple probe was also placed on the outside of the heated tube for this data set.

TABLE 1

Comparison of Particle Concentration for Particles passing (i) direct to the ASASP-X and (ii) via a normallyopen (N.O.) solenoid valve to the ASASP-X

Range 3

Channel	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	TOTAL
Direct	5368	5185	5333	6023	6138	5916	5892	5841	5644	5110	5248	5073	4841	4867	4966	81445
Through N.O. Valve	5013	4855	4897	5444	5482	5330	5205	5122	4935	4492	4547	3954	4133	4166	4203	71773
Direct	5380	5197	5318	6044	5024	5965	5851	5856	5747	5178	5340	5080	4934	4965	4970	81852

Range 2

Channel	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	TOTAL
Direct	7841	7487	6716	6755	6297	5755	5230	4495	3777	3174	2757	2348	1937	1743	1517	67829
Through N.O. Valve	6515	6220	6718	5593	5248	4795	4408	3810	3196	2674	2305	1992	1646	1456	1260	57833
Direct	7770	7446	6707	6564	6128	5594	5125	4425	3686	3123	2717	2326	1887	1702	1448	66689

Range 1

Channel	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	TOTAL
Direct	9983	5479	3897	3646	3241	1370	571	271	132	91	65	46	27	17	10	28826
Through N.O. Valve	8407	4538	3115	2882	2508	954	368	168	75	49	33	22	11	9	5.6	23145
Direct	9873	5460	3813	3584	3319	1419	580	277	134	86	65	50	28	19	12	28719

Range 0

Channel	1	2	3	4	5	6	7	8	9	10	TOTAL
Direct	8293	3336	999	320	107	47	18	5.4	2.3	1.5	13129
Through N.O. Valve	3591	1182	328	95	29	12	3.3	2.2	0.8	0.3	5244
Direct	8993	3665	1092	347	124	50	19	10	4.9	3.1	14306

TABLE 2

Comparison of Particle Concentration for Particles passing (i) direct to the ASASP-X and (ii) via a normally closed (N.C.) solenoid valve to the ASASP-X

Range 3

Channel	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	TOTAL
Direct	3114	3176	3380	4125	4402	4487	4588	4732	4788	4461	4719	4653	4663	4809	5013	65.7
Through N.C. Valve	3081	3094	3289	4001	4011	4282	4388	4534	4528	4190	4423	4398	4352	4382	4721	61787
Direct	3116	3169	3347	4143	4411	4423	4554	4634	4666	4401	4620	4630	4557	4735	4950	64370

Range 2

Channel	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	TOTAL
Direct	7241	6814	6220	6042	5655	5184	4829	4119	3564	3029	2616	2327	1959	1757	1578	62934
Through N.C. Valve	6543	6266	5713	5281	5281	4868	4533	3992	3419	2881	2550	2266	1904	1701	1590	58788
Direct	7164	6791	6203	6009	5626	5115	4726	4109	3499	2993	2591	2300	1900	1729	1570	62342

Range 1

Channel	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	TOTAL
Direct	7905	4798	3718	3888	3883	1598	898	460	286	228	180	133	86	64	51	28126
Through N.C. Valve	7440	4570	3560	3764	3547	1428	814	425	260	204	157	119	78	61	42	26430
Direct	7739	4690	3620	3779	3690	1486	862	433	248	219	176	124	73	67	49	27255

Range 0

Channel	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	
Direct	25843	10877	3940	1834	602	273	129	66	33	16	8	5	3	1.4	1.8	43515
Through N.C. Valve	20429	8603	3441	1356	536	233	113	55	31	15	8	8	2.5	1.1	1.3	33832
Direct	24666	10384	3813	1496	591	262	138	60	32	15	9	6	3	0.9	1.22	41483

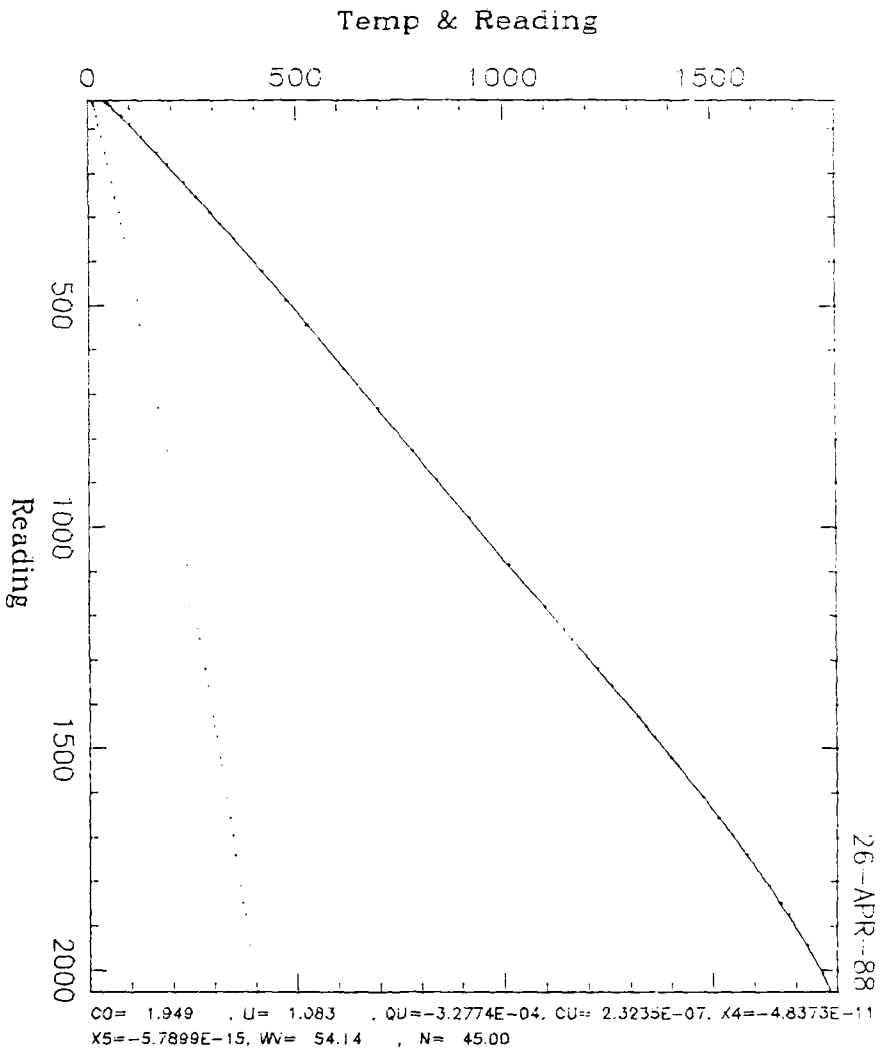

Fig. 2 A fifth order polynomial fit between the analogue readings of the thermocouple probes for the inner and outer heater tube positions.

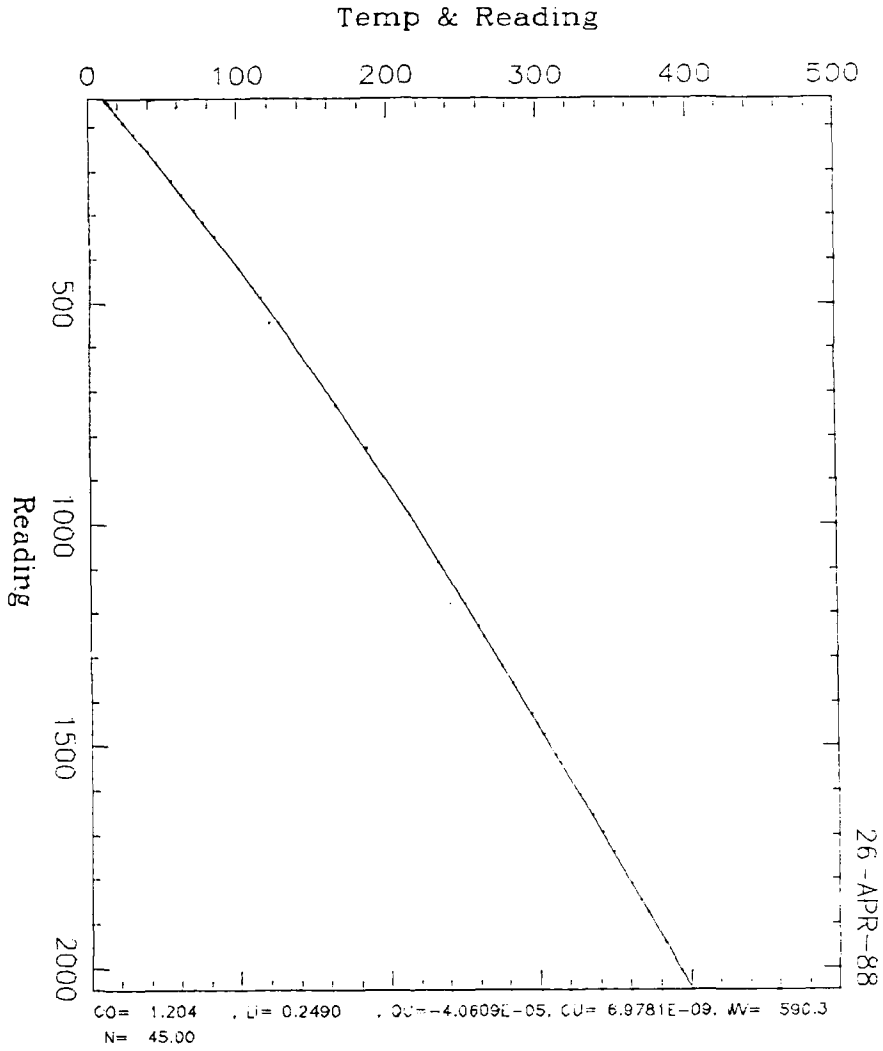

Fig. 3 A plot of the outer thermocouple analogue readings at a function of temperature.

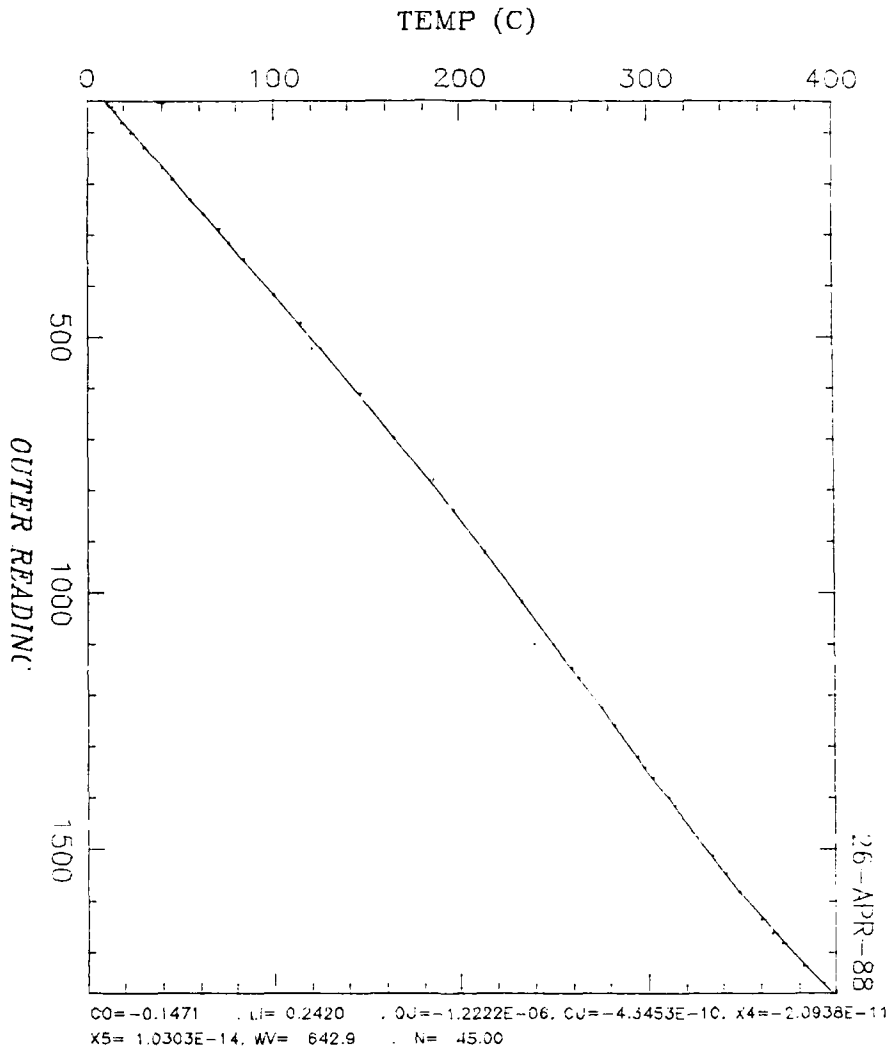

Fig. 4 A plot of the inner thermocouple analogue readings as a function of temperature.

Table 3. Field Measurement Test at Mace Head

Date	Prevaling Wind Direction	Comments
December 10 - 11, 1987	Easterly	Temperature at 300°C solenoid valves in operation 2 s data throughout
December 16 - 17, 1987	Southerly	Some measurements bypassing solenoid valves were made
January 13 - 15, 1988	South-westerly-westerly	Temperature at both 300°C and 400°C
March 10 - 12, 1988	Westerly	4 s data, temperature at 400°C; no solenoid valves in use. Temperature probe on outside of heater tube.
April 19 - 22, 1988	Southeasterly to southwesterly	No solenoid valves in use, all 400°C data.

VOLATILITY OF AEROSOLS IN THE WESTERN EUROPEAN ENVIRONMENT

The aerosol volatility apparatus was transferred to University College Galway on the basis of Government Furnished Equipment and was set up at one of the Atmospheric Sciences Research Group Labortories in late October 1987. Dr. R.G. Pinnick from the Atmospheric Sciences Laboratory, at White Sands, New Mexico, visited in late October 1987 and gave initial assistance in setting up the volatility apparatus. The apparatus was successfully set up over the month of November 1987. The Particle Measuring Systems, (PMS) Active Scattering Probe - ASASP-X (ASASP-X) probe was realigned and also calibrated for size response. In addition, the thermocouple probes were calibrated at fixed temperature reference points.

Preliminary Experimental Tests

Earlier field measurement discrepancies (R.G. Pinnick, private communication) between particle number concentration at overlapping size ranges between the two PMS probes : ASASP-X and the Classical Scattering Aerosol Spectrometer Probe (CSASP) was thought to be due to slightly different inlet temperature values for the two probes. The inlet temperature for the heater probe assembly of the ASASP-X when cooled to its lowest equilibrium values was some degrees higher than the ambient temperature. This probably caused evaporation of hygroscopic particles present, resulting in a reduction of particle number concentration compared to CSASP readings. Therefore it was decided to introduce ambient air directly to the ASASP-X through bypassing the heater intake tube section over the final time period (of order 5-7 minutes) of the heating/cooling cycle of the volatility apparatus. A schematic diagram of the apparatus used to achieve this is shown in Fig.1.

Air containing particles is drawn through a fused-quartz tube which has a nichrome ribbon wrapped tightly around it. The aerosol and air within the tube are heated to a selected temperature. A fraction of the particles evaporate and the remaining unevaporated particles or partially evaporated residue particles then pass through a normally open (N.O.) solenoid valve. The air is drawn through at a flow-rate of 5.72 $cm^3\ s^{-1}$ by means of a diaphragm pump in order to reproduce laboratory calibration flow conditions. The air flow was rendered smooth by placing a large volume buffer filter in the pump aspiration line. Particles in the line were drawn to the inlet of the ASASP-X at a calibrated flow of 1 $cm^3\ s^{-1}$ by means of the ASASP-X aspiration fan. The inter-connection tubing length which had about the same internal diameter as the quartz tubing was kept to a minimum, in order to minimize large particle losses due to sedimentation and wall losses. A normally closed (N.C.) valve was connected to the sampling tubing as shown. This valve was actuated towards the end of the heater cycle in order to allow ambient air to be sampled by the ASASP-X. The N.O. valve is actuated at the same time as is the N.C. valve which ensures that no ambient air passing through the heater section is sampled at that time.

Comparisons were made both in the field (at Mace Head Atmospheric Research Station) and in the laboratory for aerosol laden air traversing the heater tube, passing through the N.O. valve and then to the ASASP-X and ambient air being directly sampled by the ASASP-X. Preliminary checks were made to ensure that the solenoids were not leaking. Equal volumes of air were used for the two sampling lines by introducing a length of tubing in the direct path line to the ASASP-X. Laboratory generated ammonium sulphate polydispersion of particles was used as the test aerosol.

Particle concentration was recorded at 10 s intervals over a 2 minute

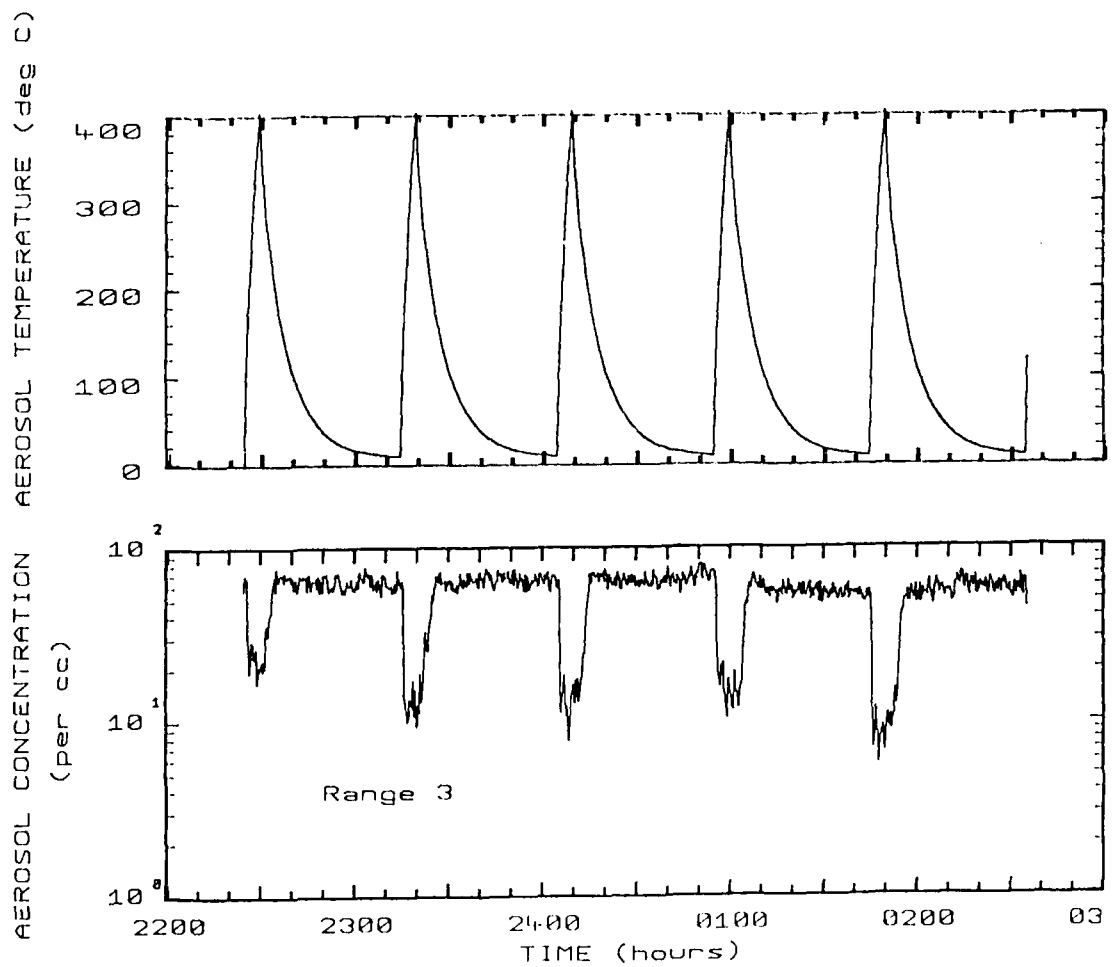

Fig. 5 Maritime aerosol volatility measurements made at Mace Head on March 10th and 11th, 1988.

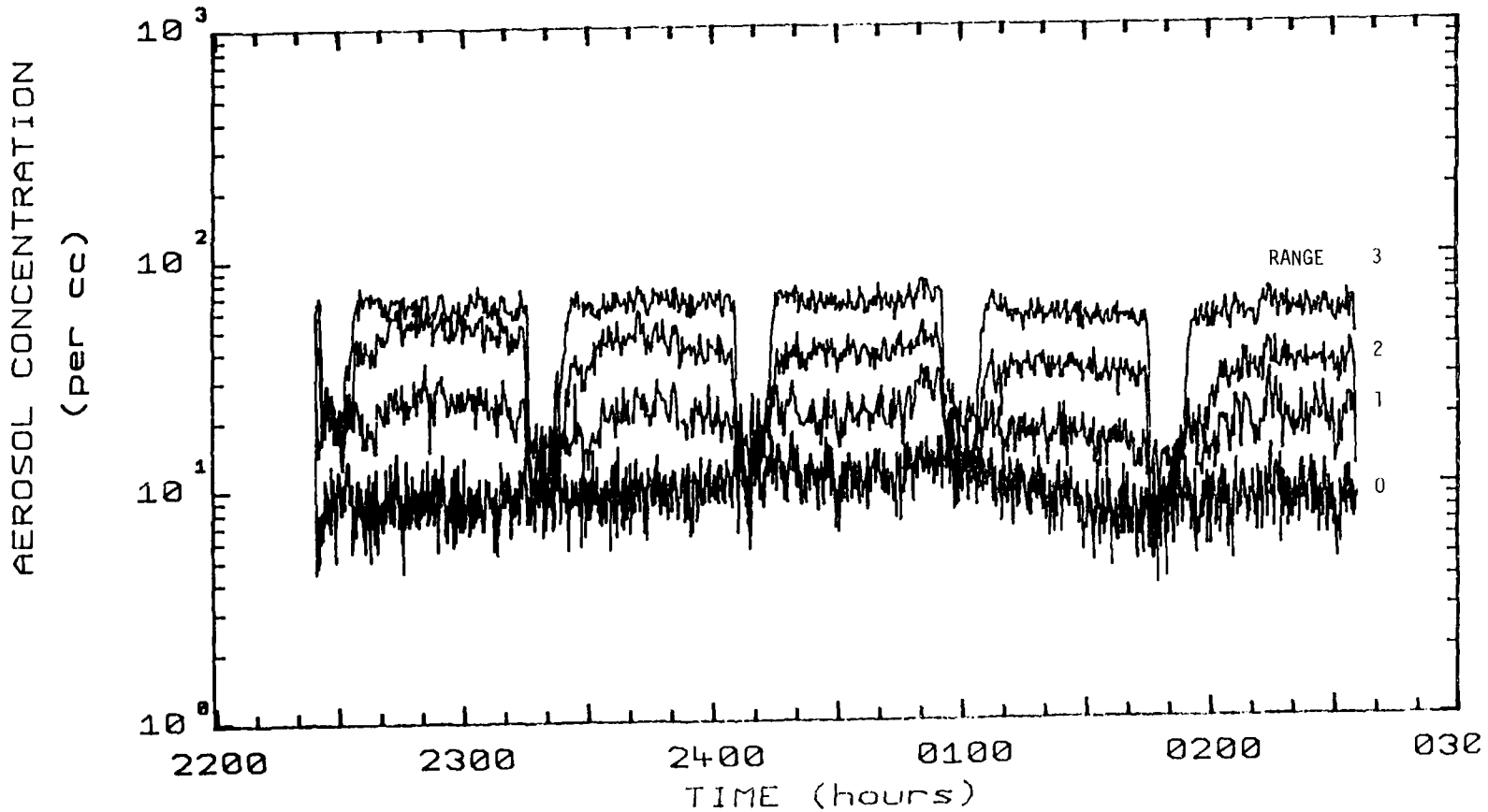

Fig. 6 Maritime aerosol volatility data for four aerosol particle ranges 0, 1, 2 and 3 (see Table 4) made at Mace Head on March 10th and 11th, 1988.

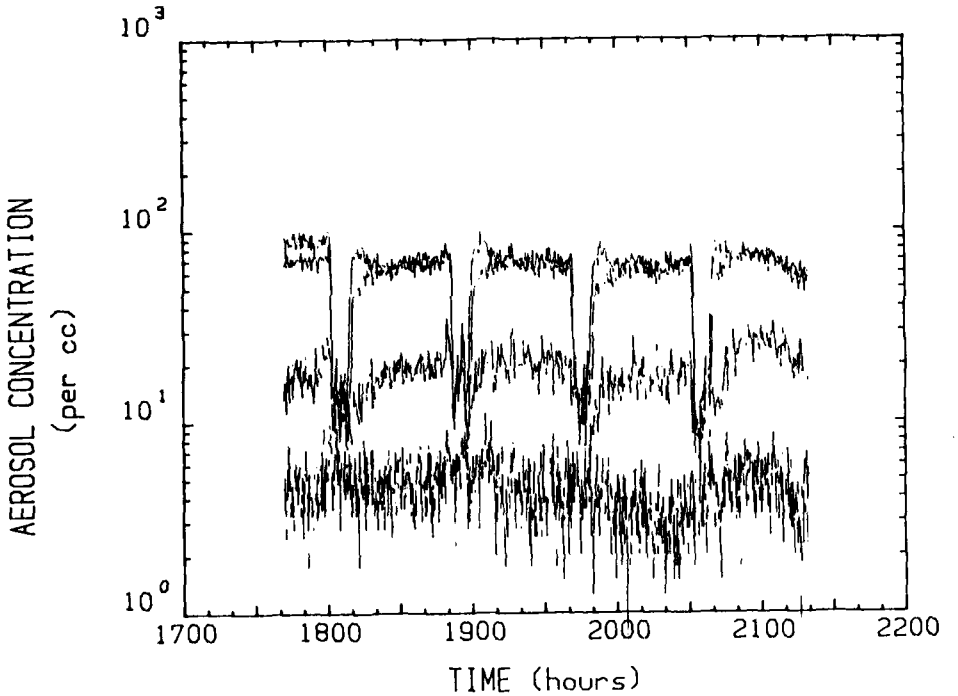

Fig. 7 Maritime aerosol volatility data for four aerosol particle ranges 0, 1, 2 and 3 (see Table 4) made at Mace Head on March 10th and 11th, 1988.

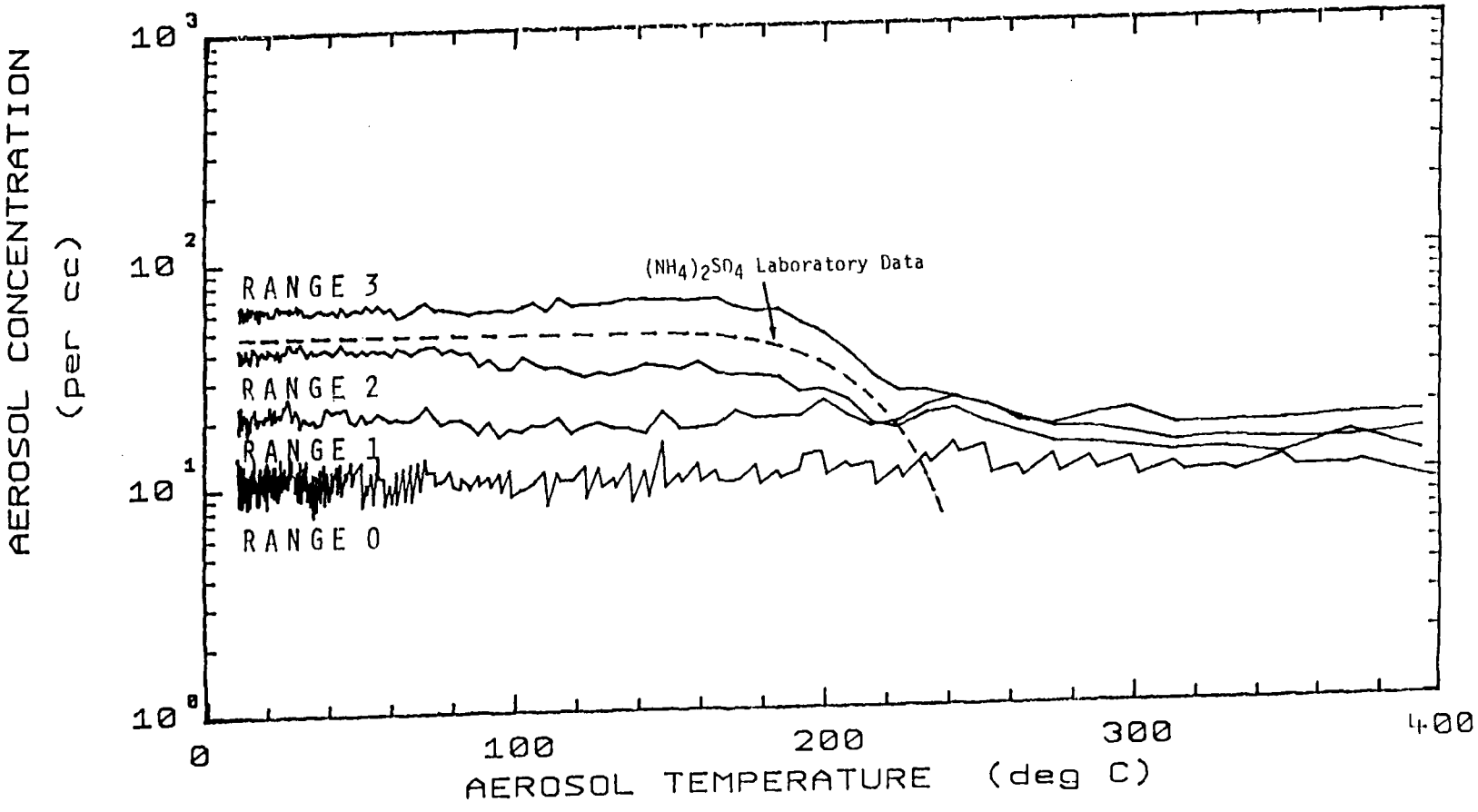

Fig. 8 Averaged temperature fractionation curves for five data cycles for particle ranges 0, 1, 2 and 3, compared to laboratory data for ammonium sulphate. The ammonium sulphate curve has been offset for ease of comparison.

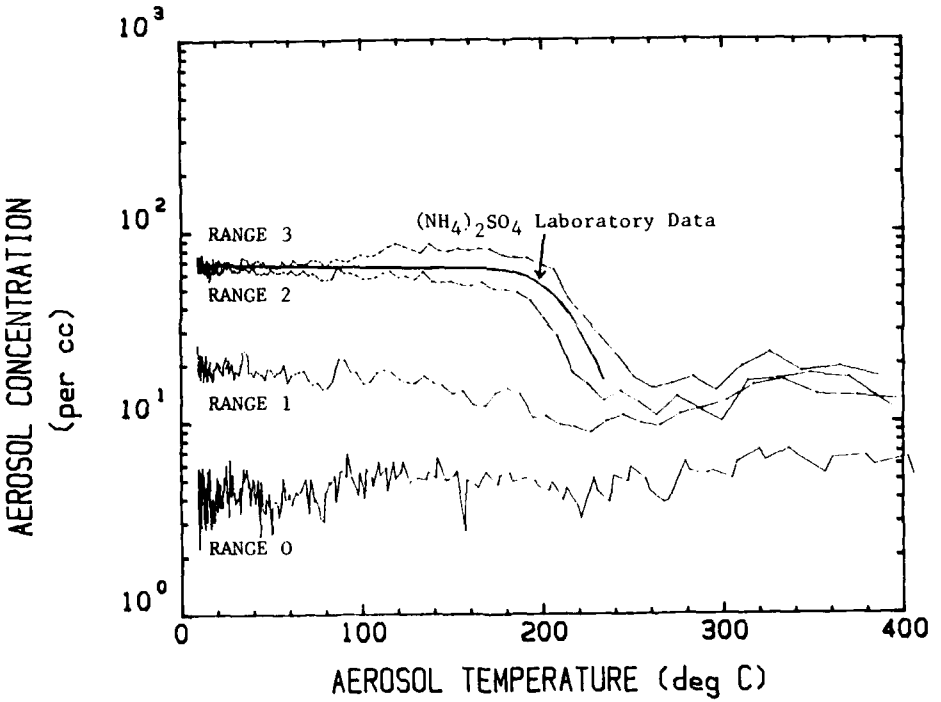

Fig. 9 Averaged temperature fractionation curves for three data cycles for particle ranges 0, 1, 2 and 3, compared to laboratory data for ammonium sulphate. The ammonium sulphate curve has been offset for ease of comparison.

Conclusions

The analysis of field meaurements of maritime aerosol to data suggests that a major constituent of the submicron fraction of the aerosol is ammonium sulphate. The next goal is to extend the temperature range of the volatility apparatus in order to determine the volatility of the supermicron fraction of the maritime aerosol.

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